

S/N: 10/510,583
Reply to Office Action of May 15, 2006

Remarks

Claims 24 - 55 are pending. Favorable reconsideration is respectfully requested.

New claims 54 and 55 have been added to more particularly point out and distinctly claim certain aspects of Applicant's invention. Claim 54 is similar to claim 24, but deletes the *Markush* group of "organohalogen compounds" (used alone) from the list of possible reactants with silicon. Claim 55 specifies that the silicon is free of catalyst, which is supported in the specification on page 4, first full paragraph. Neither claim raises any issue of new matter.

The subject invention pertains to a process for preparing silanes by reaction of silicon particles with certain reactants, in the presence of microwave energy. In most cases where they are employed, the use of microwaves is for the purpose of supplying thermal energy, instead of energy sources such as direct flames, hot air, resistance heating, heating by halogen lamps, etc. In such applications, for example the thermal cure of thermosettable coatings, the same net result is obtained, regardless of the thermal source. One would expect that to happen as well when microwave radiation is employed as an alternative to conventional sources in the reaction of silicon metal to form silanes. However, that is not the case, as more fully discussed below. Rather, exceptional yields and product specifically are observed, even when employing pure silicon, or most surprisingly, even ferrosilicon.

The claims have been rejected under 35 U.S.C. § 103(a) as unpatentable over Lewis et al. U.S. Patent 6,528,674 ("*Lewis*") in view of Griesshammer et al. Chinese patent CN 11 53138 for which an English language equivalent, CA 218 5981 is available ("*Griesshammer*"). Applicants respectfully traverse this rejection. Citations to *Griesshammer* herein pertain to the pagination of the Canadian patent.

Lewis is directed to an improvement in the preparation of the "contact mass" in the Müller-Rochow "direct synthesis" of organohalosilanes. The direct synthesis was

discovered by *Rochow*, working for General Electric, in the early 1940's.¹ Prior to the direct synthesis, organohalosilanes were prepared by a Grignard reaction, and were thus very expensive. Rochow discovered that silicon metal could be caused to react directly with halohydrocarbons in a fluidized bed at elevated temperature. However, the yield of useable products was very low unless the reaction was catalyzed by small amounts of copper. For example, as disclosed in *Rochow* U.S. Patent 2,380,995, a direct synthesis reacting methyl chloride with metallurgical grade silicon at 300°C produced only 14.5% of dimethyldichlorosilane, the desired product, while it produced 52% of the much less desired methyltrichlorosilane. Of the four possible organohalosilanes, the total yield was only about 71% based on distillate collected. The total yield based on silicon and methyl chloride is much lower. Both the yield and specificity were thus very bad. However, when copper was employed as a cocatalyst, the yield and specificity greatly improve. Unfortunately, Rochow did not report the yields using his silicon/copper contact mass, but his co-workers and numerous other researches in numerous companies did.

For example, Gilliam (General Electric) U.S. Patent 2,464,033, obtained a 44% yield of dimethyldichlorosilane (dichlorodimethylsilane) based on silicon, when a copper catalyst was employed. Moreover, of the reaction product, dichlorodimethylsilane constituted 47.3%. By also employing a zinc cocatalyst in addition to copper, yields of up to 64% dichlorodimethylsilane based on silicon used and 66% dichlorodimethylsilane based on collected product fraction were obtained. Continued improvements have steadily increased the yield and specificity, until now, yields of dichlorodimethylsilane as high as 91 - 92% are obtained. See U.S. Patent 5,817,855 (Bayer).

The mixture of silicon and copper employed in the direct synthesis is called the "contact mass" a phrase coined by the GE coworkers - see *Rochow* U.S. Patent 2,380,995, page 3, 2d column. The contact mass can be prepared in many ways, e.g. by alloying silicon and copper, by mixing particulate silicon with particulate copper, or by reaction of silicon with

¹ Independently, also, by Müller.

copper compounds such as copper oxides, copper halides, copper carboxylates, etc. *Lewis* is directed to an improvement wherein silicon and a copper compound (Cu_xO or CuCl , preferably) is mixed with silicon, and heated to a temperature less than 500°C , followed by a heat treatment at a temperature greater than 500°C . In the example, a first temperature of 350°C is employed while the second temperature is 1100°C . As a result, the specificity to dichlorodimethylsilane is increased noticeably.²

All the contact masses employed by *Lewis* are cocatalyzed and also promoted (with Zn and/or Sn as in Example 1). Only the use of alkylhalides is disclosed, specifically alkylchlorides.

Griesshammer, unlike *Lewis*, is not at all related to the direct synthesis of organohalosilanes. Rather, *Griesshammer* is directed to the reduction of tetrachlorosilane (silicon tetrachloride, SiCl_4) to trichlorosilane (HSiCl_3). The latter is used in the preparation of very pure (hyperpure) polycrystalline silicon by thermal decomposition of trichlorosilane in special batch reactors (Siemens process). The polysilicon is used as a raw ingredient for production of semiconductor materials, e.g. by the Czochralski process, especially for solar cells. Tetrachlorosilane is a reaction byproduct of thermal polysilicon synthesis. *Griesshammer* is directed to a process for reclaiming and reusing tetrachlorosilane by reducing it with hydrogen in the presence of silicon to form trichlorosilane, which can then be recycled to the polysilicon process.

To reduce the heat requirements of the process, *Griesshammer* employs microwave energy to heat the silicon particles in a fluidized bed. *Griesshammer* specifies that the silicon used is free from catalyst (*Griesshammer*, page 3, first paragraph). The reactants (H_2 and tetrachlorosilane) are incompletely reacted in the process (*Griesshammer*, p. 4), and thus the effluent gas must be separated into product (trichlorosilane) and starting material (tetrachlorosilane) and the latter recycled.

² The increase may appear to be small (In Tables 1, 2, 3, the first three entries are comparative), about 1 - 2%. However, in industry, this improvement is huge.

The combination of *Lewis* and *Griesshammer* do not teach or suggest the claimed invention for several reasons. First, there is no evidence of any motivation to one skilled in the art to combine the teachings of these references. Second, it is physically impossible to combine the references. Third, the combination, even were it proper, does not teach or suggest the claimed invention. Finally, Applicants have obtained results, even were the combination proper, which are truly surprising and unexpected.

The Court of Appeals for the Federal Circuit has spoken numerous times concerning the propriety of combining references. In the case of *In re Anita Dembiczak and Benson Zinbarg*, 50 U.S.P.Q.2d 1614 (Fed. Cir. 1999), the CAFC has indicated that the requirement for showing the teaching or motivation to combine references is "rigorous." *Dembiczak* at 1617. Moreover, this showing, which is rigorously required, must be "clear and particular." *Dembiczak* at 1617. See also, *C.R. Bard v. M3 Sys., Inc.*, 48 U.S.P.Q.2d 1225, 1232 (Fed. Cir. 1998). It is well established that merely because references can be combined, the mere suitability for logical combination does not provide motivation for the combination.

See, *Berghauser v. Dann, Comr. Pats.*, 204 U.S.P.Q. 398 (DCDC 1979); *ACS Hospital Systems, Inc. v. Montefiore Hospital*, 221 U.S.P.Q. 929 (Fed. Cir. 1984). Moreover, mere conclusory statements supporting the proposed combination, standing alone are not "evidence". *McElmurry v. Arkansas Power & Light Co.*, 27 U.S.P.Q.2d 1129, 1131 (Fed. Cir. 1993). See also, *In re Lee*, 61 U.S.P.Q. 2d 1430 (Fed. Cir. 2002).

Here, there is no evidence of any motivation to combine. In addition to this lack of evidence, which *Dembiczak* requires, the processes are entirely different and have different aims. *Lewis*'s direct synthesis is a catalyzed method for producing organohalosilanes, particularly dichlorodimethylsilane, in large quantity, at high yield, from a silicon/copper contact mass and alkylhalide. *Griesshammer*, on the other hand, does not employ the same reactants at all, as are employed in the direct synthesis. *Griesshammer* employs no alkyl halide. *Griesshammer* also employs hydrogen, which is absent in the direct process, and uses a tetrachlorosilane reactant, also absent in the direct synthesis. While the direct process is an oxidation and is highly exothermic, *Griesshammer*'s process is a reduction. While the direct

process serves to produce alkylchlorosilanes, particularly dimethyldichlorosilane, *Griesshammer's* aim is to produce trichlorosilane (HSiCl_3). One skilled in the art would not be motivated to combine *Lewis* with *Griesshammer* for any reason, and no evidence of any reason has been supplied by the Office.

Moreover, *Lewis* and *Griesshammer* are physically uncombinable and in fact diametrically opposed. *Lewis* is specifically addressed to an improved method of incorporating copper catalyst into a silicon/copper contact mass. *See, Lewis*, entire disclosure. *Griesshammer*, to the contrary, is directed to a catalyst-free process for reduction of tetrachlorosilane with silicon and hydrogen. Use of a catalyst is a salient feature of *Lewis*, while *Griesshammer* process is free of catalyst. References may not be physically combined if to do so would require ignoring salient features of the references. *In re Wesslau*, 147 USPQ 381, 393 (CCPA 1965). For either of the foregoing reasons, the rejection of the claims under 35 U.S.C. § 103(a) should be withdrawn.

Even if combined, the combination does not teach or suggest the claimed invention. The use of silicon metal (uncatalyzed), tetrachlorosilane, and hydrogen are indispensable components of *Griesshammer*. However, this combination of reactants is not one of those within the scope of the present claims. Moreover, there are numerous claims whose limitations are very far afield from any disclosure of the references, even were these references combinable. For example, claim 27 employs coarsely crystalline silicon, which is neither taught or suggested by either reference, and is generally believed to be inoperative as a reactant. Claim 28 employs amorphous silicon, not disclosed or suggested by either reference. Claim 30 requires a catalyst or promoter, necessary in *Lewis* but forbidden by *Griesshammer*. Neither reference teaches or suggests the limitation of claim 31.

Claim 32 requires a hydrogen halide. Neither reference discloses or suggests any process employing a hydrogen halide. Claim 33 requires a metal compound catalyst or promoter, forbidden by *Griesshammer*. Claim 34 is similar in this respect.

Claim 40 requires ferrosilicon. Ferrosilicon is useless in *Griesshammer* and inoperative in the Direct Synthesis. Neither reference discloses or suggests the fluorosilanes of claims 41, 49, 52, or 53. New claim 54 does not allow for the use of the reactants of either *Lewis* or *Griesshammer*, while claim 55 requires a catalyst to be present, which *Griesshammer* forbids. If the rejections of any of the foregoing claims is to be maintained, citation to the portions of the *Lewis* and *Griesshammer* references which disclose or teach the additional limitations of these claims is solicited. The claims are patentable for these reasons as well.

Finally, Applicant's process provides surprising and unexpected results. For example, in the reaction of silicon with methyl chloride as described on page 8, lines 14 and 15, a 50% yield of dichlorodimethylsilane is obtained from pure, uncatalyzed silicon (99.99% pure). By contrast, in the *Rochow* synthesis, use of pure silicon resulted in only a 14.5% yield! This result is truly surprising, and cannot be explained on the basis of a purely thermal reaction. It is cogent evidence that the use of microwaves in Applicant's process provides results which cannot be obtained thermally; the microwaves provide a distinct reaction pathway not previously available. The Example at lines 21 - 22 is even more surprising: upon dilution with argon, dichlorodimethylsilane is obtained in 87% yield, which is comparable to a catalyzed and promoted direct synthesis, yet no catalyst and no promoter is present.

In the Example at page 9, bottom to page 10, ferrosilicon is employed with methyl chloride in argon. Despite the fact that ferrosilicon is used, dichlorodimethylsilane is obtained in 82% yield, and can be increased to > 90% employing more than 1:1 methyl chloride/argon! These results indicate that a non-classical microwave effect is operative here, which may be due to creation of a silicon atomic plasma adjacent the silicon particle surface. This, then, is not merely a thermal event. The unexpected results obtained are clearly supportive of patentability.


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Applicants submit that the claims are now in condition for Allowance, and respectfully request a Notice to that effect. If the Examiner believes that further discussion will advance the prosecution of the Application, the Examiner is highly encouraged to telephone Applicants' attorney at the number given below.

Respectfully submitted,

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